SO_x-NO_x CONTROL IN A STAGED CYCLONE
COAL COMBUSTOR WITH LIMESTONE INJECTION

B. Zauderer,[1] E. Fleming[1] and W. M. Swift[2]

Coal Tech Corporation
P. O. Box 154
Merion Station, PA 19006[1]

Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439[2]

ABSTRACT

   The results of a preliminary experimental study of SO_x-NO_x control in a two-
stage cyclone coal combustor (2 MW_t) with limestone injection are presented.   The
tests were carried out using a finely pulverized West Virginia low-sulfur bituminous
coal (0.8% S), an Illinois No. 6 medium-sulfur coal (1.6% S), and an Illinois No. 6
high-sulfur coal (2.7% S). With limestone injection in either the first or second
stage of the combustor, SO_2 reductions in excess of 80% were obtained. The sorbent
injection tests indicated that the first-stage combustion temperature as influenced
by the air/fuel ratio was a major   factor in sulfur capture. NO_x emissions measured
at the exit of the second-stage combustor were reduced by up to 55 % over those from
single-stage combustion. In addition, the benefits of a staged cyclone combustor
for NO_x control were enhanced when the second-stage gas temperature was minimized,
either through increasing second-stage air dilution or by maximizing first-stage
carbon conversion.

BACKGROUND AND OBJECTIVES

   Recent developments in coal cleaning technology and in the preparation of coal-
water slurries have prompted renewed interest in the direct firing of gas turbines
with coal-derived fuels, such as partially cleaned fuel gas and coal-water mixtures.
Several recent studies have identified the potential economic advantages of these
systems over conventional coal-fired power generation (1-3).

   An alternative approach to the concept of a coal-fired gas turbine is the direct
combustion of the coal in a staged slagging combustor. The slag rejecting combustor
has been developed primarily in support of magnetohydrodynamics (MHD) technology.
More recently, however, development of the slagging combustor for the control of SO_x
and NO_x emissions has been considered for application to both the coal-fired gas
turbine option and to the retrofit of existing gas and oil-fired boilers.

   The concept of the staged slagging combustor for SO_x/NO_x control is similar to
the LIMB (limestone injection into multi-stage burners) concept being developed for
conventional boilers by the U. S. Environmental Protection Agency and the Electric
Power Research Institute: namely, staged combustion (fuel-rich first stage) for the
control of NO_x emissions and limestone injection for the control of SO_x emissions.
The added advantage of slag rejection during combustion makes the slagging combustor
ideally suited to a wide range of utility and industrial applications, including the
coal-fired gas turbine system and gas/oil-to-coal conversions.

   Although the amount of data available in the literature is fairly limited, the
application of advanced slagging combustors to the control of SO_x/NO_x emissions has
shown promising results (4,5).   In pilot-scale testing of a low NO_x/SO_x burner
developed by Rockwell International, better than 70% SO_2 reduction, NO_x emissions
below 100 ppm (at 3% O_2), slag/fly ash removal over 70%, and carbon burnout greater
than 99% have been demonstrated in tests with low-sulfur western coals (4). In a
few cases, better than 95% SO_x reduction and zero NO_x emissions were reported, even
when the only calcium used was the calcium inherent in the coal ash.

Equally encouraging results have been reported for the TRW slagging combustor developed in part for MHD applications (5). SO_2 removals of between 50 and 90% are claimed in limestone injection tests at a Ca/S ratio of 3 for coals with sulfur contents ranging from 0.6 to 2.1%. NO_x emissions have ranged from 330 to 450 ppm (at 3% O_2) and carbon conversions were in excess of 99%.

Because of the proprietary aspects of the staged slagging combustor tests just described, details concerning them and the operational parameters that influence the control of SO_2 and NO_x emissions have not been reported. Hence, a series of combustion tests with limestone injection was undertaken in a nominal 2 MW staged slagging combustor at Argonne National Laboratory (ANL). The objective of the tests was to investigate the control of SO_x and NO_x emissions at conditions simulating direct coal firing into a gas turbine.

<center>EXPERIMENTAL</center>

Facility

The test program was conducted in the ANL Fossil Energy Users Laboratory (FEUL), which is a DOE-owned facility constructed to accommodate a wide range of experiments related to coal and oil combustion. The facility has two test legs: one with a combustor capable of burning a variety of liquid fuels (including slurry mixtures) and the other equipped with the two-stage slagging coal combustor used in performing the present work. Each test leg has the necessary independent fuel-feed, support, and control systems. The air compressors, combustion air preheater, effluent control equipment, coolant system, and data acquisition system are shared.

The coal combustor, designed and fabricated by ANL, is illustrated in Fig. 1. The combustor consists of three flanged sections with a fourth section (not shown) downstream of the combustor for instrumentation and gas sampling. The primary combustor consists of two sections: the first section accommodates the combustion air inlet header, coal feed injection nozzle, and a limestone feed port; the second section accommodates a slag tap. Constructed from 24-in. (0.64-m) schedule 20 carbon steel pipe, the primary combustor is refractory lined to a nominal inside diameter of 0.55 m. The overall length is 1.37 m.

As illustrated in Fig. 1, part of the primary combustion air enters axially along the combustor centerline and the remainder enters either tangentially (louvers installed) or radially (louvers removed). The coal to the first stage also enters along the combustor centerline axis. A pintle with an angle of 45° is located at the end of the coal injection line to radially disperse the incoming coal. First-stage limestone injection was accomplished through a port on the front face of the combustor and at an angle that intersected the centerline of the combustor at a point approximately 0.4 m downstream of the coal injector nozzle.

The second stage of the combustor is constructed from 18-in. (0.46-m) schedule standard pipe and is refractory lined to a nominal inside diameter of 0.38 m. The secondary combustion air enters radially through three ports located around the combustor wall. During certain tests, the limestone sorbent was injected through a port just upstream of the secondary air inlet nozzles. The second stage of the combustor is separated from the first stage by a slag baffle with a centerline opening of 0.25 m.

The coal combustor has a nominal thermal input rating of 2 MW, including the sensible heat of the combustion air, which can be preheated up to 1090 K. For the tests that form the subject of this paper, the thermal input to the combustor was in the range 1.1 to 2.4 MW, with most of the tests in the lower end of this range. The split between the primary and secondary combustion air and the ratio of primary axial to swirl air can be independently controlled. For all but one of the tests described herein, the louvers in the primary combustor were not installed; hence, the nonaxial primary combustion air was injected with little or no swirl component.

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Test Coals and Limestone

Three different coals were used in the test program: a West Virginia Alma C seam coal and two Illinois No. 6 seam coals. The results from nominal analyses of the three coals are given in Table 1. The West Virginia coal was primarily selected because nearly 70% of the total sulfur in the coal (0.85 wt %) is organic. It was speculated that this might ensure rapid release of the sulfur from the coal during fuel-rich combustion in the first stage, possibly enhancing the capture of the sulfur by the limestone sorbent. The two Illinois coals were selected because of their higher total sulfur (1.6 and 2.7 wt %) and ash content. A limitation on the limestone hopper capacity and injection rate made testing with a very high sulfur coal (>3 wt %) impractical.

All three coals were delivered to a commercial grinding service where the coals were pulverized and loaded into metal bins with a nominal capacity of 900 kg. The first two coals were pulverized to approximately 98% -200 mesh. The third coal was slightly coarser at 92% -200 mesh. The bins of coal were delivered to ANL and, as needed, automatically loaded into a coal feed storage hopper capable of holding approximately 11,400 kg, a quantity sufficient for combustion tests of 6 to 8 hours.

The limestone used in this work was obtained from Grove Lime Co. in Stephens City, Virginia. This limestone, which contains 95.3 wt % $CaCO_3$ and 1.3 wt % $MgCO_3$, was used extensively in the early development of fluidized-bed combustion technology. For this study, the limestone was pulverized to 80% -200 mesh.

Test Procedure and Conditions

The general procedure adopted for each test was to bring the combustor to the desired initial set of operating conditions over a period of one to two hours. Baseline gas analysis data were then recorded without limestone injection. Limestone would then be injected into the first and/or second stage of the combustor and the composition of the gas exiting the second stage of the combustor would be monitored for changing SO_2 concentration until a new "steady-state" value of SO_2 was achieved. The limestone feed would then be discontinued and the SO_2 level in the combustion gas allowed to return to the baseline value. A change in operating

Table 1. Nominal Analyses of Test Coals (Dry Basis)

	Coal		
Analysis	West Virginia Alma C Seam	Illinois No. 6 (Orient #4 Mine)	Illinois No. 6 (Fidelity #11 Mine)
Test Coal Designation	1	2	3
Moisture, As Received	1.2	4.7	7.0
Proximate Analysis, wt %			
Ash	6.0	9.2	12.3
Volatile Matter	33.8	39.5	35.4
Fixed Carbon	60.2	51.3	52.2
Sulfur	0.85	1.6	2.7
Ultimate Analysis, wt %			
Carbon	81.5	73.9	70.7
Hydrogen	5.2	5.7	4.9
Nitrogen	1.4	1.5	1.4
Sulfur	0.85	1.6	2.7
Ash	6.0	9.2	12.3
Oxygen	5.1	8.2	8.0

parameters would then be made and the above procedure repeated at the new set of conditions. This procedure would generally be repeated three or four times during each combustor test period to obtain data over a range of conditions.

Gas analysis probes were located in the first and second stages of the combustor and also downstream of the second stage. The gas samples were cooled as they were withdrawn from the sampling ports and passed through a gas conditioning system where the sample was filtered free of particulates and dried using a membrane-type dryer. The samples then passed to on-line gas analyzers for CO, O_2, SO_2, CO_2, and NO/NO_x analysis. The analyzers were calibrated using standard calibration gases at the beginning of each test and at frequent intervals during each test period.

The nominal test conditions and the measured reductions in SO_2 leaving the second stage of the combustion system are listed in Table 2. The parameters varied in the course of this study were the primary and overall air/fuel ratios (stoichiometry), first- and/or second-stage limestone injection, Ca/S molar ratio, and sulfur content of the coal.

<center>RESULTS</center>

Combustor Operation

Although the ANL combustor performed very well during the limestone injection tests, the combustor did have a number of performance characteristics that influenced the test program. The ANL combustor, for example, is limited to atmospheric pressure operation, whereas gas turbine applications would require

<center>Table 2. Nominal Test Conditions and Experimental Results</center>

Test Condition	Coal[a]	Coal Feed Rate (kg/h)	Stoichiometry Primary	Overall	Limestone Injection Ca/S	Stage	Sulfur Retention (%)
2.3-2	1	225	0.5	1.2	3.2	1	23
3.1-2	1	145	0.6	1.7	2.3	1	36
3.1-3	1	145	0.6	1.7	4.4	2	97
4.1-2	1	150	0.7	1.7	2.5	1	49
4.1-3	1	150	0.7	1.7	2.5	2	26
4.2-2	1	150	0.6	1.7	1.0	1	33
5.1-2	2	165	0.9	1.5	3.1	2	49
5.2-2	2	200	0.4	1.2	2.5	1	24
5.3-2	2	140	0.6	1.7	3.6	2	55
5.4-2	2	110	0.9	2.2	4.9	2	31
6.1-2	2	145	1.1	1.7	3.5	2	62
6.2-2	2	135	1.2	1.8	3.6	2	51
6.4-2	2	155	0.6	1.6	2.6	2	72
6.5-2	2	145	0.6	1.7	3.1	2	84
7.1-2	3	170	1.2	1.6	2.6	1	50
7.2-2	3	155	1.2	1.7	2.7	1	44
7.4-2	3	155	0.8	1.7	2.8	2	58
7.5-2	3	155	0.8	1.8	2.9	1	48
8.1-2	3	185	0.6	1.6	2.3	1	55
8.2-2	3	155	0.6	1.7	2.6	1	46

[a] Refer to Table 1 for coal designations

operation at elevated pressure. This was not considered a serious limitation, however, because the identification of parameters affecting SO_x/NO_x control at atmospheric pressure would generally be useful in any application of the concept.

Also, the available supply of combustion air was insufficient to operate the combustor at high thermal inputs and maintain the high excess air conditions of 180 to 200% required to simulate gas turbine applications. As a result, heat losses to the combustor walls were as high as 15 to 20% of the thermal input. This, together with occasional flameouts of the combustor, resulted in slag solidifying and blocking the slag drain in the first stage of the combustor. As a result, the slag rejection capabilities of the ANL combustor could not be evaluated. Further, it was not possible to determine the quantity of slag and limestone that accumulated in the combustor over the duration of a test. Hence, accurate carbon and sulfur material balances at the individual test conditions could not be determined.

A third factor in the tests, which was not a limitation of the equipment, was the decision to operate the combustor without the swirl louvers in the primary combustor. As a result, the combustor performed essentially as a plug flow unit with respect to both gases and particles. In this respect, the ANL combustor more nearly resembled the Rockwell low NO_x/SO_x burner cited above.

The high particle loadings and slagging conditions in the primary combustor also made sampling of the first-stage combustion gases very unreliable. Sampling ports would quickly slag over and/or sampling lines plug, making further sampling impossible. While gas sampling lines at the exit of the second stage of the combustor were also prone to plugging, the availability of multiple sampling ports and the ability to periodically clean lines not in use made second-stage gas sampling highly reliable. Hence, the evaluation of combustor performance was largely based on the results of the second-stage gas analysis.

Evaluation of Limestone Injection for Sulfur Control

As shown in Table 2, measured reductions in SO_2 emissions ranged from a low of 23% to a high of 97% over the full range of test conditions investigated, including both first- and second-stage sorbent injection. The test results indicate that sulfur capture via limestone injection is a complex phenomenon. Since, however, the air/fuel ratio in the primary combustor has a significant influence on both (1) the rate of sulfur release from the coal and the ratio of H_2S to SO_2 formed during fuel-rich combustion, and (2) the resulting first- and second-stage gas temperatures, it was found that the SO_2 reduction during sorbent injection exhibited a noticeable dependency on the first-stage air/fuel ratio.

Figure 2 is a plot of the effect of first-stage combustion stoichiometry on reducing SO_2 during first-stage limestone injection. In producing the plot, the sulfur reduction data in Table 2 were normalized to a Ca/S ratio of 3.0 assuming a linear dependency. Figure 2 also includes some data from injection tests with a hydrated limestone in a related series of tests. The results indicate the optimum reduction in SO_2 occurred at an air/fuel stoichiometric ratio in the range 0.5 to 0.7. This is considered to be largely a thermal effect in that higher first-stage air/fuel ratios resulted in excessively high first-stage gas temperatures (>1900 K, calculated). The apparent low reduction in SO_2 during first-stage injection at an air/fuel ratio of 0.4 is not fully understood.

Figure 3 is a similar plot of SO_2 reduction as a function of first-stage air/fuel stoichiometry for second-stage sorbent injection. The observed dependency is generally the same as for first-stage sorbent injection. This may have resulted from the selection of the second-stage sorbent injection location, which was slightly upstream of the secondary air injection. Hence, the first-stage gas temperatures may also have influenced sorbent performance during second-stage injection.

Although the data are fairly limited and exhibit a high degree of scatter, the general results are encouraging in that the overall measured sulfur reductions in excess of 80% at Ca/S ratios on the order of three are consistent with the results reported for the Rockwell and TRW combustors. Considerably more data are needed, however, to fully understand the complex effects of operating parameters on combustor performance.

NO_x Control

Independent of coal type, the staged combustion for NO_x control exhibited two different dependencies on the first-stage air/fuel ratio, as shown in Fig. 4. The upper curve, which corresponds to less reduction in NO_x emissions, is representative of tests where the second-stage bulk gas temperature was too high (>1750 K) and/or where carbon conversion in the first stage was relatively low (<70%), leading to excess carryover of fuel-bound nitrogen in the char to the oxidizing second stage of the combustor. Greater NO_x reductions occurred when either the second-stage temperatures were lower than 1650 K and where carbon conversion in the fuel-rich first stage was high enough (>70%) to allow release and conversion of the fuel bound nitrogen to N_2.

In both cases, however, measured second-stage NO_x was reducible to less than 100 ppmv at 15% O_2 by controlling the first-stage stoichiometry. This corresponds to 300 ppmv at 3% O_2, a value that is again consistent with the results reported by TRW and Rockwell for their combustors.

CONCLUSIONS

The results of the preliminary investigation of limestone injection in a staged slagging combustor for the control of SO_x and NO_x emissions are very encouraging. Sulfur retentions were in excess of 80% and NO_x emissions were as low as 100 ppmv (at 15% O_2). Considerably more testing is required, however, to fully understand the complex mechanisms controlling the sulfur retention process. The results are consistent with those reported for the TRW and Rockwell slagging combustors.

ACKNOWLEDGMENTS

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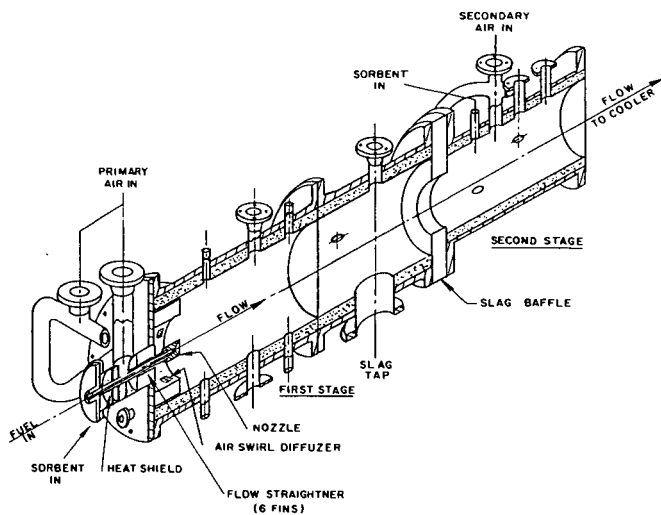
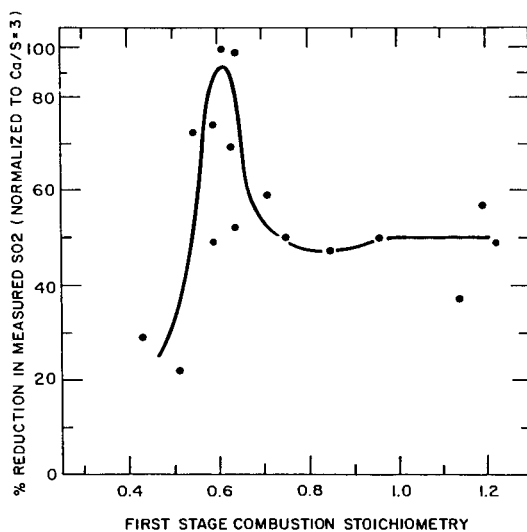Fig. 1.   Schematic of the ANL 2-Stage Slagging Combustor

Fig. 2.   Measured Reduction in SO_2 with First Stage
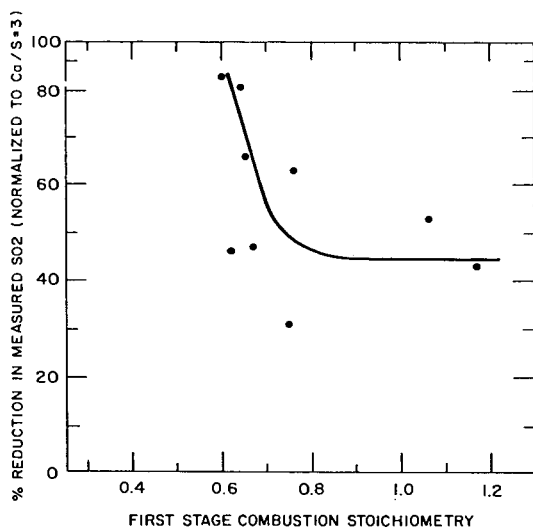   Limestone Injection

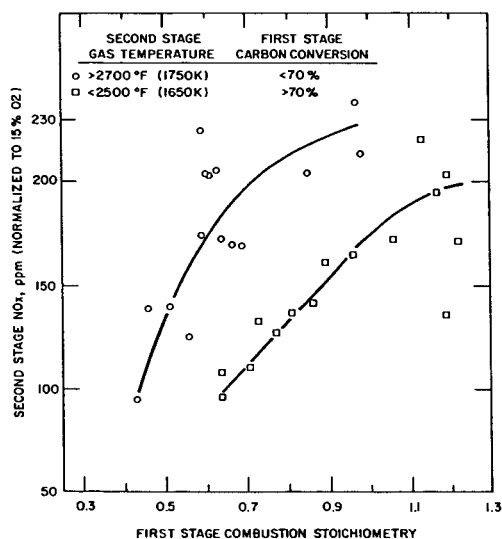Fig. 3. Measured Reduction in SO_2 with Second Stage
Limestone Injection

Fig. 4. Measured NO_x Emissions during Limestone
Injection Tests